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(54) Title: ADHESIVE COMPOSITION AND METHOD FOR ADHERING TEXTILES TO EPDM RUBBER

(57) Abstract: An improved adhesive composition and method for adhering textile reinforcing elements to rubber; particularly ethylene-propylene-diene rubber in the manufacture of rubber articles such as power transmission belts, wherein said composition comprises a latex of a hydrogenated styrene-butadiene rubber, a hydrogenated nitrile-butadiene rubber, a carboxylated hydrogenated nitrile-butadiene rubber, an ethylene-propylene rubber, a chlorosulfonated polyethylene or blends thereof; an aqueous solution of a maleinized liquid polybutadiene; and, optionally, up to about 10 % by weight carbon black dispersion.

ADHESIVE COMPOSITION AND METHOD FOR ADHERING TEXTILES TO EPDM RUBBER

Technical Field

The present invention relates to the bonding of textile reinforcements to high temperature resistant rubber and, more particularly, this invention relates to an improved adhesive composition for adhering such textile reinforcements to EPDM (Ethylene-Propylene-Diene rubber) for use in reinforced rubber based products such as high temperature resistant power transmission belts.

Background Art

With the increased demand for higher temperature power transmission belts, there is a need for an improved adhesive to bond textile reinforcements to rubber compositions used in the manufacture of high temperature resistant articles. In bonding rubber to a reinforcing material in products such as power transmission belts and other reinforced high temperature resistant rubber products, the common practice is to treat the material with a RFL latex dip to enhance the bonding of the rubber to the material.

U.S. Patent No. 3,325,333 to Kigane et al. teaches a method of adhering a polyester cord to a CSM rubber compound by treating the cord with an aqueous adhesive composition containing methylolated blocked organic polyisocyanate and a vulcanizable organic polymer at conventional or normal polyester treating temperatures.

U.S. Patent No. 3,060,078 to Atwell teaches a method of bonding polyester cord fibers to a CSM rubber compound by treating the cord with a resorcinol-formaldehyde neoprene latex adhesive composition

Typically, the RFL dip is prepared by mixing an RFL prepolymer with a latex. The RFL prepolymer provides adhesion to the reinforcing material and the latex provides adhesion to the rubber. Where

the surface reactivity of the reinforcing material is low, as is the case with polyester and aramid cords, the cord is usually pre-treated with a composition which improves its reactivity. Compositions that have been used for this purpose include epoxy resins and isocyanates. Conventionally, belts are prepared by bonding a double-twisted cord to the rubber. The cord is immersed in a RFL dip and transported through a vertical oven where it is heated under tension for a predetermined period of time. This causes the cord to elongate and restructure the polyester. This process is known as "stress-elongation" or "heat set". The process dries the adhesives which are coated on the cord. The common RFL dip systems have also been developed for general purpose rubber compounds like natural rubber and styrene-butadiene-rubber. For these rubbers, the latex used in the RFL dip is often a terpolymer based on styrene, butadiene and vinylpyridine. For less reactive rubber compounds, like EPDM, the common RFL dip systems are not suitable.

Prior art methods of adhering polyester cord (for example, to rubbers such as chlorosulfonated polyethylene (CSM) have included the use of neoprene latex and vinyl pyridene latex in the RFL latex dip.

Another conventional treatment for polyester cord is taught in U.S. Pat. Nos. 54,654,099 and 5,807,634, wherein the polyester cord receives an initial polyphenylisocyanate treatment at 8 to 15 pounds of tension which is beat activated at a temperature in the range of 300° F to 425° F for 120 seconds to react the functional groups of the polyphenylisocyanate with the open bond sites in the cord. A RFL adhesive is coated over the reacted polyphenylisocyanate and dried, preferably in an oven, for 120 seconds at a temperature in the range of 180° F to 275° F to evaporate the water from the RFL and to keep the RFL from blistering prior to the heat setting of the cord. The heat setting of the polyester cord at 460° F for 60 seconds takes place after the adhesives are applied and therefore, the adhesives are subjected to this elevated temperature. Adhesion between RFL treated textiles and EPDM

rubbers can be achieved if the EPDM rubber is compounded in such a way as to allow for proper reaction chemistry (crosslinking) to occur at the interface of the rubber and adhesive. Generally, these modifications to the EPDM rubber reduce the desired physical properties and dynamic flex life of the rubber compound. Accordingly, there is a need for an improved adhesive that is able to adhere textiles to an EPDM rubber that is compounded for extended belt life and temperature resistance.

Disclosure of the Invention

The present invention relates to an adhesive composition which, when applied to a textile reinforcement using conventional methods in an environmentally safe manner, provides effective adhesion of the textile reinforcement to a rubber compound, and more particularly EPDM, and also relates to a method for adhering such textiles to rubber compounds.

In the first embodiment of the present invention, the adhesive composition comprises the following: (a) a latex of hydrogenated styrene-butadiene rubber (HSBR), hydrogenated nitrile-butadiene rubber (HNBR), carboxylated hydrogenated nitrile-butadiene rubber (XHNBR), EPDM (ethylene-propylene-diene), or chlorosulfonated polyethylene (CSM), blended with (b) an aqueous solution of maleinized liquid polybutadiene. The adhesive composition may also utilize a combination of any of the latices listed above, and may further contain latex blends with one or more latices of the following: SBR (styrene butadiene), nitrile, vinyl pyridine, natural rubber, acrylonitrile, EVA, PVA; polyester such as ethylene vinyl acetate (EVA), polyvinyl acetate (PVA), etc; polyurethane, PVC, polychloroprene , acrylic acid, methacrylic acid, vinylidine chloride, butyl, and copolymers and terpolymers thereof. It has been discovered that this combination of ingredients results in a composition that attains excellent adhesion of textiles to rubber compound formulations. The adhesive composition exhibits excellent hot

durability and cold flex properties while maintaining the adhesive bond between the textile – rubber interface.

In the second embodiment of the invention of the method for adhering reinforcing cord to the rubber compounds comprises:

dipping the cord into an adhesive composition comprising: (a) a latex hydrogenated styrene-butadiene rubber (HSBR), hydrogenated nitrile-butadiene rubber (HNBR), carboxylated hydrogenated nitrile-butadiene rubber (XHNBR), EPDM (ethylene-propylene-diene) or blends thereof, and (b) an aqueous solution of maleinized liquid polybutadiene;

drying the adhesive;

incorporating the cord into the rubber compound; and

curing the cord and rubber compound to produce power transmission belts.

Detailed Description of Preferred Embodiments

In accordance with the present invention, therein is provided an improved adhesive that will adhere ethylene-propylene-diene rubber to a reinforcing textile with excellent performance at both high and low temperatures. Traditionally, the adhesive system is applied to the reinforcing textile in a fluid carrier, either aqueous or solvent, dried, and heat set to achieve specific properties in the textile. The improved adhesion between the EPDM rubber and the reinforcing textile, in accordance with the invention, provides unexpected good results with respect to adhesion force between the EPDM rubber and the reinforcing fiber as well as improved rubber tear.

The adhesive used in the present invention comprises (a) a latex such as hydrogenated styrene-butadiene rubber (HSBR), hydrogenated nitrile-butadiene rubber (EPDM), chlorosulfonated polyethylene (CMS), carboxylated chlorosulfonated polyethylene (ACSM) and the like, blended with (b) an aqueous solution of malienized liquid polybutadiene. The adhesive composition may contain a combination of

the above noted latices and may also contain latex blends of one or more styrene-butadiene rubber (SBR), nitrile rubber, vinyl pyridine, natural rubber, acrylonitrile, ethylene-vinyl acetate (EVA), polyvinyl acetate (PVA), polyester, polyurethane, polyvinyl chloride (PVC), polychloroprene, acrylic acid, methacrylic acid, vinylidene chloride, butyl rubber, and copolymers, terpolymers and mixtures thereof. It has been found that the present combination of ingredients results in an improved adhesive composition that provides excellent adhesion of textiles to EDPM rubber compounds. Preferably, the latex used in the present invention is hydrogenerated styrene-butadiene rubber (HNBR), carboxylated hydrogenated nitrile-butadiene rubber (XHNBR), or chlorosulfonated polyethylene (CSM) having a solids content of about 25 to 50%. Most preferably, the latex has a solids content of about 32 to 45%. The latex content of the adhesive is preferably about 50 to 90 percent by weight and preferably about 65 to 90 percent by weight.

The resin utilized in the present invention is typically an aqueous maleinized polybutadiene; more specifically it is an ammonia neutralized isobutyl half-ester of maleinized liquid polybutadiene. Typically, the malienized liquid polybutadiene is prepared by reacting about 5 to 25 parts of maleic anhydride with 100 parts of liquid polybutadiene (Lithene). The amount of malienized liquid polybutadiene utilized in the adhesive composition of the present invention ranges from about 1 to 50%, preferably about 10 to 30% by weight. The concentration of the maleinized liquid polybutadiene in the aqueous solution as defined in the present invention is the concentration of the "half-ester" which typically is present in amounts of about 15 to 30% by weight and preferably about 19 to 25% by weight.

For the purpose of this invention, the malienized liquid polybutadiene is defined as an aqueous solution of malienized liquid polybutadiene although it typically is not a true solution until after hydrolysis and neutralization to the isobutyl half ester. Water, preferably

de-ionized water, is utilized in combination with the essential components of the present invention in order to provide an adhesive composition having a final solids content of about 5 to 40 percent, preferably about 15 to 30 percent.

The adhesive composition of the present invention may further contain up to about 10% by weight carbon black dispersion. While the solids content of the carbon black dispersion is not believed to be critical, the solids content is typically about 30 to 50% and even more typically about 35 to 40% carbon black in water.

The latex of the present invention may be further blended with up to a major amount of one or more additional latices of the following rubbers: styrene-butadiene, nitrile, vinyl pyridine, natural rubber, acrylonitrile, ethylene-vinyl acetate, polyvinyl alcohol, polyurethane, polyvinyl chloride, polychloroprene, acrylic acid, methacrylic acid, vinylidine chloride, butyl, and copolymers and terpolymers thereof. Typically, up to about 75% by weight of these additional latices based upon the total weight of latices can be employed. The additional latices have a solids content of about 30 to 50%.

After the adhesive is applied to the textile either by dipping, spraying, or brushing, preferably dipping, it is then dried in an oven at a temperature in the range of about 82.2 to 135° C (180° F to 275° F), preferably about 107.2 to 121.1° C (225° F to 250° F) for an effective time, typically, about 2 minutes. Finally, the textile is then heat set in an oven for about 1 to 3 minutes at a temperature in the range of from 148.9 to 232.2° C (300° F to 450° F), preferably 176.7 to 204.4°C (350°F to 400°F) under a pull tension specific to textile type, denier, twist multiplier, and or weave to attain specified tensile member properties. The adhesive composition of the present invention can optionally contain other well known additives including plasticizers, fillers, pigments, thickeners, dispersing and wetting agents, reinforcing agents and the like, in amounts employed by those skilled in the adhesive arts to obtain the

desired consistency, appearance, reinforcement, and uniformity of coating on the textile substrate.

The reinforcing textile of the present invention can be any of the various textiles conventionally employed in forming textile reinforced rubber products, particularly, power transmission belts. For example, exemplary textiles include polyamide fiber; meta-and para-aramide fiber; polyester fiber such as PET (polyethylene terephthalate), PVA (polyvinyl alcohol), and the like; PEN (polyethylene naphthalate) fiber, cotton, fiber, carbon fiber, glass fiber, PBO (poly p-phenylene-2,6-bezobisoxazole) fiber, acrylic fiber; rayon fiber; LCP (liquid crystal polymer) fiber; and the like. Preferably, the reinforcing element is a polyester.

It is desirable that the adhesive-containing reinforcing textile be incorporated into the EPDM rubber compounds as individual cords and then cured in a conventional manner to produce rubber articles such as power transmission belts, more specifically Polyrib belts, synchronous belts, variable speed belts, flat belts, raw edge v-belts, and wrapped V-belts. The reinforcement element may also be in the form of an adhesive-containing fiber sheet between two or more layers of the EPDM rubber. In another aspect of the invention, the adhesive-containing fiber is chopped into short strands and dispersed throughout the rubber article. Any one or a combination of the above may be used to reinforce the rubber article.

As a result of low surface reactivity of the polyester, it may be desirable to pretreat the textile reinforcing element with a pre-dip composition in order to enhance the surface reactivity of the polyester cord. Such pretreatment is discussed in commonly assigned U.S. Pat. No. 5,654,009 and 5,807,634, the contents of which are incorporated herein by reference to the extent of the pretreatment.

For a more complete understanding of the invention, the following examples are presented to show specific uses of the techniques

taught.

Example 1

A mixture of 70 grams hydrogenated styrene-butadiene rubber latex (32% solids)[1], 10 grams carbon black dispersion (37.5% solids)[2], and 20 grams ammonia-neutralized aqueous solution of isobutyl half-ester of maleinized liquid polybutadiene (21.6% solids)[3], was diluted to a final solid content of 23% and coated onto an isocyanate pretreated polyester cord by dip application at 4% dip pick up. The treated cord was dried in a 107.2°C (225°F) oven for 2 minutes and then heat set under 10 lbs. of tension 204.4°C (400°F) for 1 minute. After heat setting, samples were prepared according to ASTM D1871, Method B, except that the cord was wound in closely spaced fashion against a first ply of EPDM rubber across the width of a mandrel. Samples of cord sandwich were then cut from the mandrel and tested according to Method B. Results were recorded as peak peel strength (measured in pounds per inch of width) and percent rubber retention at the rubber/treated cord interface. Results are shown in Table 1.

Table 1

Test Condition	#Peel Per ASTM D1871, Method B (LBS)	%Rubber Retention (Estimated Percent rubber retention on 1 inch wide Sample)
22.2°C (72°F) Primary adhesion	73	100
121.1°C (250°F) Hot Box (30 min soak)	24	100
-40°C (-40°F) Cold box (30 min soak)	81	100

Example 2

Example 1 was repeated using 60 grams of the hydrogenated styrene-butadiene rubber, 10 grams of the carbon black dispersion, and 30 grams of the ammonia-neutralized aqueous solution of

isobutyl half-ester of maleinized liquid polybutadiene. The results are shown in Table 2.

Table 2

Test Condition	#Peel Per ASTM D1871, Method B (LBS)	%Rubber Retention (Estimated Percent rubber retention on 1 inch wide Sample)
72°F Primary adhesion	50	100
250°F Hot Box (30 min soak)	21	100
-40°F Cold box (30 min soak)	65	90

Example 3

Example 1 was repeated using 70 grams of hydrogenated nitrile-butadiene rubber latex (45% solids)[4], 10 grams carbon black dispersion (37.5% solids), and 20 grams ammonia-neutralized aqueous solution of isobutyl half-ester of maleinized liquid polybutadiene (21.6% solids). The results are shown in Table 3.

Table 3

Test Condition	#Peel Per ASTM D1871, Method B (LBS)	%Rubber Retention (Estimated Percent rubber retention on 1 inch wide Sample)
72°F Primary adhesion	45	75
250°F Hot Box (30 min soak)	18	100
-40°F Cold box (30 min soak)	49	40

Example 4

Example 1 was repeated using 70 grams carboxylated hydrogenated nitrile-butadiene rubber latex (40% solids)[5], 10 grams carbon black dispersion (37.5% solids), and 20 grams ammonia-

neutralized aqueous solution of isobutyl half-ester of maleinized liquid polybutadiene (26.6% solids). The results are shown in Table 4.

Table 4

Test Condition	#Peel Per ASTM D1871, Method B (LBS)	%Rubber Retention (Estimated Percent rubber retention on 1 inch wide Sample)
72°F Primary adhesion	48	95
250°F Hot Box (30 min soak)	20	100
-40°F Cold box (30 min soak)	48	50

Example 5

Example 1 was repeated using 70 grams chlorosulfonated polyethylene (40% solids)[8], 10 grams carbon black dispersion (37.5% solids), and 20 grams ammonia-neutralized aqueous solution of isobutyl half-ester of maleinized liquid polybutadiene (21.6% solids). The results are shown in Table 5.

Table 5

Test Condition	#Peel Per ASTM D1871, Method B (LBS)	%Rubber Retention (Estimated Percent rubber retention on 1 inch wide Sample)
72°F Primary adhesion	36	40
250°F Hot Box (30 min soak)	18	90
-40°F Cold box (30 min soak)	31	20

(1) Goodyear chemical HSBR Latex

(2) Solution dispersion Ajax black 36

(3) Revertex Lithene n4-5000

(4) Sumitomo Seika HNBR Latex

(5) Zeon chemicals zetpol B Latex

(6) Sumitomo Seika CSM 450 Latex

Having described the invention in detail and by reference to the preferred embodiments thereof, it will be apparent that modifications and variations are possible without departing from the scope of the invention defined in the appended claims.

The present invention is, of course, in no way restricted to the specific disclosure of the specification and drawings, but also encompasses any modifications within the scope of the appended claims.

CLAIMS:

1. An adhesive composition for use in bonding a textile reinforcing element to rubber, said adhesive composition being characterized by:

(a) a latex of a hydrogenated styrene-butadiene rubber, a hydrogenated nitrile-butadiene rubber, a carboxylated hydrogenated nitrile-butadiene rubber, and ethylene-propylene-diene rubber, a chlorosulfonated polyethylene, or blends thereof; and

(b) an aqueous solution of a maleinized liquid polybutadiene.

2. The adhesive composition of claim 1, characterized in that said composition comprises about 50 to 90% by weight of said latex, said latex having a solids content of about 25 to 50%, and/or in that said composition comprises about 1 to 50% by weight aqueous solution of said maleinized liquid polybutadiene, wherein said aqueous solution of maleinized liquid polybutadiene can be an aqueous solution containing about 19 to 25 percent by weight isobutyl half-ester of maleinized liquid polybutadiene, wherein said aqueous solution of isobutyl half-ester of maleinized liquid polybutadiene can be neutralized, for example with ammonia.

3. The adhesive composition of claim 1, characterized in that said composition further comprises up to about 10% by weight carbon black dispersion, said dispersion having a solid content of about 30 to 50%.

4. The adhesive composition of claim 1, characterized in that said latex is further blended with up to about 75% by weight of a second latex having a solids content of about 30 to 50% of said second latex comprising styrene-butadiene rubber, a nitrile rubber, polyvinyl pyridine, natural rubber, polyacrylonitrile, polyester, polyurethane,

polyvinyl chloride, polychloroprene, polyacrylic acid, polymethacrylic acid, polyvinylidine chloride, butyl rubber, copolymers or terpolymers thereof, or mixtures thereof, based upon the total weight of latex.

5. The adhesive composition of claim 1, characterized in that said omposition has a solids content of about 5 to 40%, especially a solids content of about 25 to 30%.

6. The adhesive composition of claim 1, characterized in that said textile reinforcing element is selected from the group consisting of polyamide fiber, meta- and para-aramid fiber, polyester fiber, polyethylene naphthalate fiber, cotton fiber, carbon fiber, glass fiber, poly [p-phenylene - 2,6-benzobisoxazole] fiber, acrylic fiber, rayon fiber, and liquid crystal polymer fiber, especially polyester fiber, and/or in that said rubber is ethylene-propylene-diene rubber.

7. An adhesive composition having a solids content of about 5 to 40% for use in bonding a polyester reinforcing element to an EPDM rubber compound in the manufacture of a power transmission belt, said adhesive composition being characterized by:

(a) about 50 to 99% by weight of a latex of a hydrogenated styrene-butadiene rubber, a hydrogenated nitrile-butadiene rubber, an alkylated hydrogenated nitrile-butadiene rubber, or a chlorosulfonated polyethylene, said latex having a solid content of about 25 to 50%;

(b) about 1 to 50% by weight of any ammonia-neutralized aqueous solution of an isobutyl half-ester of maleinized iquid polybutadiene wherein said aqueous solution contains about 15 to 30% by weight half-ester of maleinized liquid polybutadiene; and

(c) up to about 10% by weight carbon black dispersion, said dispersion having a solids content of about 30 to 50%.

8. A method for improving the adhesion of a textile

reinforcing element to a rubber compound, characterized by the steps of:

applying an adhesive to said textile reinforcing element, said adhesive comprising: (a) about 50 to 99 weight percent of a latex of a hydrogenated styrene-butadiene rubber, a hydrogenated nitrile-butadiene rubber, a carboxylated hydrogenated nitrile-butadiene rubber, an ethylene-propylene-diene rubber, a chlorosulfonated polyethylene, or blends thereof, said latex having a solids content of about 25 to 50%; and (b) about 1 to 50 weight percent of an aqueous solution of a maleinized liquid polybutadiene wherein said aqueous solution contains about 15 to 30% by weight half-ester of maleinized polybutadiene;

drying the adhesive containing textile reinforcing element at a temperature of about 82.2 to 135°C;

heat setting the dried adhesive containing textile reinforcing element at a temperature of about 148.9 to 232.3°C;

incorporating the textile reinforcing element having the adhesive applied thereto, into a high temperature resistant rubber compound to provide a reinforced high temperature resistant rubber article; and

curing said rubber.

9. The method of claim 8, characterized in that said adhesive comprises a blend of:

(a) about 65 to 90% by weight of said latex of a hydrogenated styrene-butadiene rubber, a hydrogenated nitrile-butadiene rubber, a carboxylated hydrogenated nitrile-butadiene rubber, or a chlorosulfonated polyethylene;

(b) about 10 to 30% by weight of said ammonium neutralized aqueous solution of an isobutyl half-ester of maleinized liquid polybutadiene; and

(c) up to about 10% by weight carbon black dispersion, said dispersion having a solids content of about 30 to 50%.

10. The method of claim 8, characterized in that said textile reinforcing element is selected from the group consisting of polyamide, meta- and para-aramid, polyester, polyethylene naphthalate, cotton fiber, carbon fiber, glass fiber, poly (p-phenylene-2,6-bezobisoxazole), acrylic fiber, methacrylic fiber, rayon fiber, and liquid crystal fiber, especially a polyester, and/or in that said high temperature resistant rubber is ethylene-propylene-diene rubber, and/or in that said reinforced high temperature resistant rubber article is a power transmission belt.

INTERNATIONAL SEARCH REPORT

International application No.
PCT/US01/40327

A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) :C09J 5/02
US CL :156/307.5, 910

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 156/307.5, 910; 524/501

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	JP 4-81476 A (KIYOMITSU) 16 MARCH 1992 ENGLISH TRANSLATION ABSTRACT	1-10
Y	GB 2,042,563 A (NEUBERT) 24 SEPTEMBER 1980 ABSTRACT, PAGE 1 LINES 5-9, PAGE 3 LINES 12-44, PAGE 4 LINES 79-102	1-10
Y	US 3,515,616 A (MIYAMOTO etal) 02 JUNE 1970 ABSTRACT, COL 1 LINES 25-27, COL 3 LINES 1-4, COL 4 LINES 58-67, COL 5 LINE 16 THRU COL 6 LINE 2	1-10
Y,P	US 6,127,476 A (JIALANELLA etal) 03 OCTOBER 2000 ABSTRACT, COL 3 LINES 28-32	1-10

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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"O"	document referring to an oral disclosure, use, exhibition or other means		
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